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TRITIUM BODY BURDEN OF CHILDREN 1967-1968

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Tritium Body Burden of Children, 1967-1968

A. A. Moghissi and *Robert Lieberman*[1]

The tritium concentration in body water of children was determined from samples of urine taken from two children in each of nine Institutional Total Diet Sampling stations. The tritium concentrations in body water of children gradually decreased from about 1.5 nCi/liter to less than 0.2 nCi/liter during the period, January 1967 to December 1968. Several anomalies were noted.

The dose equivalent from tritium for standard man was calculated to be 0.15 mrem for 1967 and 0.13 mrem for 1968. The dose equivalent for children varies according to age and is roughly 0.1 mrem for a 13-year-old child.

After the cessation of atmospheric nuclear testing, tritium concentration in the environment has continuously decreased. Tritium is, however, produced in nuclear reactors and is eventually introduced into the hydrosphere. The Bureau of Radiological Health (BRH) has recognized the importance of this radionuclide and has established a number of networks to evaluate a possible radiation hazard to the public. Analytical results of surface water networks (*1*) are continuously reported. Similarly, rain is continuously monitored for tritium (*2*).

The ultimate purpose of most networks is the evaluation of the radiation dose or, to be more explicit, the dose equivalent (DE) to man. The most suitable medium for the radiobioassay of tritium is body water. It is well known that tritiated water introduced into the body is evenly distributed in the body water within a short period of time. (*3*).

The body burden may thus be expressed as the concentration of tritium in the body water, and urine may be used for radiobioassay and the evaluation of DE. In connection with the Institutional Total Diet Sampling Network Program (ITDSNP) (*4*), urine samples from children in several children's homes were analyzed for a number of radionuclides at the Southeastern Radiological Health Laboratory. During the 1967–1968 period a portion of these samples was radioassayed for tritium.

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Sampling and analysis procedures

Samples were collected from 9 ITDSNP stations distributed throughout the continental United States. The locations of these stations are shown in figure 1. To obtain a reasonable sampling accuracy, samples were collected from two children from each home. Samples were collected in disposable containers and an aqueous (1 ml) solution of merthiolate was used as a preservative. Upon arrival at the laboratory, the samples were logged and stored in a cold room at a temperature of 0-4° C.

Approximately 30 ml of the urine were distilled in all glass equipment using potassium persulfate as an oxidizing agent (*5*). Tritium assay was carried out by liquid scintillation counting, using a dioxane-based scintillation liquid (*5*). Twenty ml of dioxane, containing 7 g of 2,5 diphenyloxazole (PPO) and 1.5 g of p-bis-(methylstyryl)-benzene (bis-MSB) in 1 liter, was mixed with 5 ml of distilled urine and counted in a Beckman liquid scintillation counter. Duplicate samples were prepared and each was counted for 50 minutes twice. A background sample was counted prior to the first sample and a second background sample counted after the second replicates. If any one of the 50-minute sample counts deviated by more than 2 σ from the average, the duplicates were recounted. The minimum limit of detection (MLD) of this system at 2 σ confidence level was 0.2 nCi/liter. Beginning with the July 1968 samples, one of the replicates was counted using a suspension technique (*6*). A p-xylene solution containing 7 g

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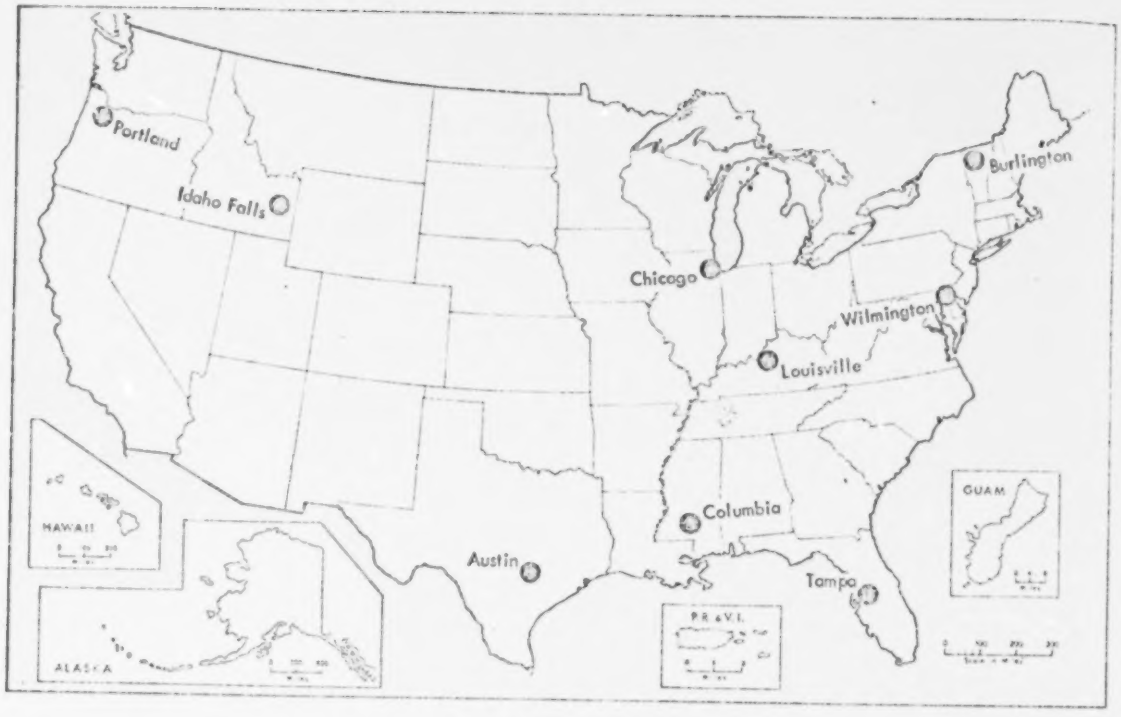

Figure 1. Nine Institutional Total Diet Sampling stations)

PPO and 1.5 g bis-MSB/liter was mixed with a detergent, Triton N101 (a product of Rohm and Haas, Philadelphia, Pa.), in a volume ratio of 2.75:1. Fifteen ml of xylene-detergent mixture and 10 ml of water were thoroughly mixed and counted. The details of this procedure are described by Lieberman and Moghissi (*6*). The minimum limit of detection of this suspension technique was 0.1 nCi/liter.

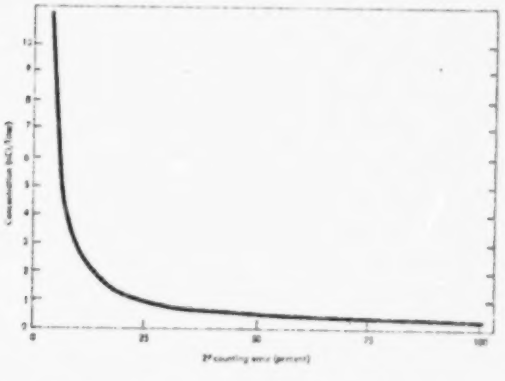

Figure 2. Correlation between the tritium concentration of water and the counting error in percent at 2-sigma confidence level

Although the samples counted during the latter part of 1968 had a lower minimum limit of detection, for the sake of uniformity and because the new procedure was in an experimental state, 0.2 nCi/liter is used as the MLD for the entire period. Figure 2 shows the counting error at 95-percent confidence level for the activity range of this investigation.

Results

From tables 1 and 2, it may be seen that tritium concentrations in the body water of children have gradually decreased in some cases, from approximately 1.5 nCi/liter to less than 0.2 nCi/liter from January 1967 to December 1968.

Certain anomalies are, however, striking. In several cases, the two samples collected from children in the same home show considerable discrepancy. Originally, this was attributed to analytical inaccuracies. In all these cases, however, a rerun of the samples confirmed the original findings.

In search for a reason, a number of possibilities were considered. The water or food intake was the first possibility. It is, however, highly improbable

Table 1. Tritium concentration in urine, 1967

Location	Concentration (nCi/liter)											
	January	February	March	April	May	June	July	August	September	October	November	December
Vt: Burlington	1.8 1.7	1.5 1.4	1.4 1.5	1.5 NS	1.0 .7	0.5 .9	NS NS	NS NS	NS NS	NS NS	NS NS	NS NS
Del: Wilmington	1.2 1.3	.6 1.3	.8 .8	.5 .4	NS .4	.5 .4	0.4 .6	1.3 1.4	0.4 NS	0.3 .3	NS NS	0.3 .2
Ky: Louisville	4.0 1.8	3.0 1.3	3.2 1.1	1.0 1.0	1.8 .5	.8 .4	1.2 .8	2.8 1.8	.9 .8	NS NS	.5 .6	NS NS
Fla: Tampa	1.0 1.0	NS NS	.5 .4	.5 .7	< .2 < .2	< .2 < .2	< .2 < .2	< .2 < .2	.4 3.0	NS NS	NS NS	< .2 < .2
Miss: Columbia	1.6 1.2	.4 .5	.7 .6	< .2	.3 .3	< .2 < .2	< .2 < .2	.8 .8	< .2 < .2	< .2 < .2	.2 .4	< .2 < .2
Ill: Chicago	1.6 1.3	.6 NS	.6 .6	.7 .6	.7 .5	.3 .5	.4 .4	1.2 .8	NS NS	.4 .4	NS NS	< .2 < .2
Ore: Portland	.8 NS	1.2 NS	1.2 1.0	.6 .5	.4 .3	.3 .3	NS NS	NS NS	.4 .2	< .2	NS NS	NS NS
Tex: Austin	1.3 1.2	.9 .8	.9 .9	.9 .6	.3 .2	.3 < .2	.4 NS	.8 6.0	.6 4.9	.4 NS	.3 .4	< .2 .3
Idaho: Idaho Falls	1.4 1.3	NS NS	1.3 1.0	1.0 1.0	.4 .8	.5 .7	1.0 .8	12.0 .9	.4 NS	.2 .6	NS NS	.5 3.8

NS, no sample.

Table 2. Tritium concentration in urine, 1968

Location*	Concentration (nCi/liter)											
	January	February	March	April	May	June	July	August	September	October	November	December
Del: Wilmington	0.2 < .2	≤0.2 < .2	0.5 1.0	0.5 < .2	0.2 .5	1.9 1.2	0.4 < .2	0.9 .8	0.2 .4	≤0.2 < .2	0.2 .2	7.2 .4
Ky: Louisville	.6 .4	.5 .9	.2 NS	.5	.4	.8 .4	.3 NS	.3 NS	.3 NS	1.4 NS	.3 NS	.5
Fla: Tampa	1.5 9.6	< .2 < .2	NS NS	< .2 .4	< .2 < .2	.2 .2	< .2 < .2	< .2 < .2	.4 .8	< .2 < .2	< .2 NS	.4 NS
Miss: Columbia	.5 < .2	.4 2.2	2.0 < .2	.2 < .2	2.4 .4	.4 4.0	< .2 < .2	< .2 .4	1.0 .7	< .2 < .2	< .2 NS	< .2 < .2
Ill: Chicago	.2 5.7	.9 2.0	< .2 4.0	2.0 .9	.4 < .2	NS NS	NS NS	.4 .4	NS NS	NS NS	NS NS	NS NS
Ore: Portland	< .2 < .2	NS NS	.6 < .2	6.0 < .2	.2 .2	.6 .4	< .2 < .2	< .2 < .2	1.0 .8	< .2 .6	< .2 < .2	6.0 < .2
Tex: Austin	NS NS	< .2 < .2	< .2 < .2	< .2 < .2	< .2 < .2	.2 .4	.4 .4	< .2 < .2	< .2 < .2	1.0 < .2	< .2 < .2	.2 .8
Idaho: Idaho Falls	5.0 4.7	.9 2.0	1.4 .9	.8 .6	.5 .7	.5 .5	1.0 .8	NS NS	1.0 1.0	.5 < .2	.6 .8	1.0 1.0

* No samples were received from Burlington, Vt., during 1968
NS, no sample received.

that a person living in Tampa, Fla., would have a tritium body burden of 9.6 or 4.5 nCi/liter (January 1968) as water collected in that area had a tritium concentration of less than 0.2 nCi/liter. Tritium concentration in food collected during the same month was less than 0.2 nCi/liter (7) of water of combustion.

It is also unusual that two children from Wilmington, Del., having essentially the same tritium intake, show differences of almost 7 nCi/liter (December 1968).

A second possibility is the contamination of the sample during the sample collection and analysis. This possibility was systematically investigated. Forty-five disposable containers randomly selected from a variety of shipments were filled with 1 liter of water and left in the cold room for 14 days. These water samples were subse-

quently processed in the same manner as the urine samples and counted. No case of increased activity could be observed.

The only other possibility was the contamination of the samples during the shipment. One characteristic of the samples showing anomalies was their condition on arrival. They were generally spoiled, presumably due either to improper preservation or because the container was washed prior to sample collection, thus removing the preservative.

It is known that the specific activity of hydrogen (*8*) and methane (*9*) in the atmosphere is several orders of magnitude higher than that of atmospheric moisture. If during the biochemical reactions that occur during spoiling of urine samples, methane becomes incorporated into the aqueous portion of the urine, then an increased specific activity may result. The true reason for the relatively high specific activities found remains unknown for the time being. Further investigations are necessary to clarify this phenomenon.

Dose calculations

Moghissi and Porter (*1*) proposed the following equation for the calculation of tritium dose under equilibrium conditions:

$$DE = 10^{-4}C(QF)$$

where,

DE = dose equivalent in mrem/a;
C = concentration of tritium in the body in nCi/liter; and,

QF = quality factor for tritium.

This equation assumes a body weight of 70 kg of which 10 percent is hydrogen. To facilitate the dose calculation, it is assumed that the hydrogen concentration of the body is essentailly independent of age. This assumption is substantiated by the findings of Forbes (*10*) who has shown that the body water to body weight ratio changes only slightly with age. He also determined the body weight of children as shown in table 3. As the sex of the children was not recorded, the average weights were used in the calculations.

Since the publication of the above equation, Evans (*11*) has found that under continuous exposure, the specific activity of organically-bou[illegible] and aqueous hydrogen is essentially the same. This finding confirms the underlying idea leading to that equation and is in conflict with the recommendations of ICRP (*12*).

Numerous investigations have been published recently dealing with the quality factor (QF) of tritium. Feinendegen (*13*) has reviewed the literature extensively. Vennart (*14*) in a recent review has proposed a QF of 1. It is unfortunate that none of these papers reviews Shtukkenberg's (*15*) paper where a QF of 3 was proposed. Although Lambert (*16*) found recently that a QF of 1.3–2.4 was more appropriate, the International Commission on Radiological Protection decided to follow Vennart's recommendation (*17*) and has proposed a QF of 1 for tritium. Since no definite decision has been made by the National Commission on Radiation Protection and Measurements, a QF of 1.7 was found to be appropriate and is used in the calculations.

To evaluate the dose, all values of less than 0.2 nCi/liter are assumed to be 0.1 nCi/liter. This

Table 3. Body weight of children

Age (years)	Weight (kg)		
	Male	Female	Average
9	30.5	32	31.25
10	32	35	33.5
11	37	38	37.5
12	43.5	41	42.25
13	45	48	46.5
14	51	56	53.5

Table 4. Average tritium concentrations in urine of children, total body burden, and dose equivalents for the standard man

Location	1967			1968		
	Concentration in urine (nCi/liter)	Total body burden (nCi)	Dose equivalent (mrem)	Concentration in urine (nCi/liter)	Total body burden (nCi)	Dose equivalent (mrem)
Vt: Burlington	1.26	79.4	0.21			
Del: Wilmington	.67	42.2	.11	0.72	45.4	0.12
Ky: Louisville	1.46	92.0	.25	.56	35.3	.10
Fla: Tampa	.48	30.2	.08	.89	56.1	.15
Miss: Columbia	.40	25.2	.07	.68	42.8	.12
Ill: Chicago	.62	39.1	.11	1.42	89.5	.24
Ore: Portland	.34	21.0	.09	.79	49.8	.13
Tex: Austin	1.02	64.3	.17	.22	13.9	.04
Idaho: Idaho Falls	1.30	94.5	.26	1.30	75.6	.20
Average	0.88	55.6	0.15	0.81	51.0	0.13

value is estimated to be the current tritium level in surface waters of low specific tritium activity. Also high values are used in the averaging process although there is a probability that the calculated dose would be somewhat too high.

Table 4 shows the average tritium concentrations in the body water of children at the various sampling stations. The total body burdens are calculated for standard man, assuming that the tritium concentration in the body of standard man would be essentially the same as that in the bodies of children. The DE is calculated also for standard man and is included in the tables. The DE for standard man during 1967 was 0.15 mrem and during 1968, 0.13 mrem.

Table 5 shows the average total body burden and the DE for various ages calculated for the average tritium concentration.

Table 5. Tritium total body burden (TBB) and the dose equivalents (DE) of children during 1967 and 1968

Age (years)	Total body burden (nCi)		Dose equivalents (mrem)	
	1967	1968	1967	1968
9	24.8	22.8	0.067	0.061
10	26.6	24.4	.071	.066
11	29.8	27.3	.080	.074
12	33.5	30.8	.090	.083
13	36.9	33.8	.099	.091
14	42.5	38.9	.114	.105

The DE for children varies according to age and is roughly 0.1 mrem for a 13-year-old child.

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